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Optics

High-coherent oscillations in IR spectra of macroporous silicon with nanocoatings

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Abstract. IR light absorption oscillations in 2D macroporous silicon with CdTe, ZnO and CdS surface nanocrystals, microporous and SiO_2 layers were compared taking into account the electro-optical Wannier–Stark effect. We proposed a high-coherent optical quantum computer based on ZnO nanoparticles on macroporous silicon surface with the broadening parameter of the Wannier–Stark steps of 10^{-3} and the IR oscillation coherence of 0.25–0.4% at room temperature. This value is much less than the coherence of cold atoms (5%) and diamond crystal with the nitrogen impurity (10–17%). A logical qubit of this quantum computer is the presence/absence of the infinite resonance scattering of oscillated electron by the Wannier levels in the electric field at the silicon matrix – nanocoating boundary. The scattering amplitudes are controlled and measured at room temperature by using the resonant maxima of IR absorption.

Keywords: macroporous silicon with nanocoatings, infinite resonance scattering, optical quantum computer.

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1. Introduction

2D macroporous silicon is a promising material for silicon photonics and nanoelectronics, since it provides preparation of structures with the required geometry and large effective surface. It is related with electro-optical effects in macroporous silicon: (1) the Franz–Keldysh electro-optical effect [1] in the direct band-to-band optical range, (2) the linear impurity Franz–Keldysh effect [2] for the weak electric field approximation, and (3) the Wannier–Stark electro-optical effect in the mid-infrared range (MIR) at the strong electric field approximation [3].

2D macroporous silicon structures show the Franz–Keldysh oscillations caused by the intrinsic electric field on the macropore surface with 1.7-nm depth [1]. In view of the potential barrier on the macropore surface, one should take into account recharging of the local surface centers at the energies below that of the indirect band-to-band transition. The experimental MIR absorption spectra of macroporous silicon agree well with the corresponding spectral dependences of the electro-optical energy and the imaginary part of permittivity in the weak electric field approximation thus confirming realization of the impurity Franz–Keldysh effect [2]. In addition, we investigated the MIR light absorption oscillations in 2D macroporous silicon with surface nanocrystals and SiO_2 layers, taking into account the electro-optical effects at strong electric fields. The resonant electron scattering by surface bonds and realization of the Wannier–Stark effect were confirmed [3, 4]. In this case, the Wannier–Stark effect is related with the large-time electron scattering as compared with the period of absorption oscillations in the strong electric field of the illuminated "silicon – nanocoating" boundary.

In this paper, we have compared the MIR light absorption oscillations in 2D macroporous silicon with CdTe, ZnO and CdS surface nanocrystals, microporous and SiO_2 layers with account of the electro-optical Wannier–Stark effect. We have analyzed the shift and deviations of oscillation peaks; the broadening parameter Γ of the Wannier–Stark steps; the influence of "quantum superiority" on coherence of the Wannier levels. In addition, we have proposed the high-coherent optical quantum computer based on a silicon matrix with macropores and a layer of nanocrystals on the surface of macropores for implementation of the Wannier–Stark quantum electro-optical effect.

2. Procedure

The samples to be studied were made of silicon wafers with the thickness $H = 520$ μm, resistivity of 4.5 Ohm·cm, characterized by the [100] orientation and *n*-type of conductivity (the electron concentration $n_0 = 10^{15}$ cm^{-3}). We used the technique of electrochemical etching under backside illumination of the silicon substrate [5]. Macropores were etched in the form of a square lattice of parallel air cylinders with the diameter $D_p = 2 \pm 0.2$ μm, period 4 μm, depth $h_p = 50…100$ μm and concentration $N_p = 6.25 \cdot 10^6$ cm^{-2} (Fig. 1a). The initial specimens are complex micropore-macropore silicon structures consisting of 100-nm micropore layers on macropore walls. Additional anisotropy etching in 10% solution of KOH permits to remove microporous layers from the macropore surface.

CdTe nanocrystals of 20 nm in size were deposited on macroporous silicon substrates (Fig. 1b) by modified installation of metal dispersion using the "hot wall" molecular epitaxy [6]. The undoped CdTe crystal sputtered at the substrate temperature 475 K and source temperature 650 K served as a material for evaporation. The thickness of films (200 nm) was set by time of structure staying above the evaporation source [7].

Methods for synthesizing ZnO nanoparticles in isopropanol and from solution of zinc compound of $Zn(CH_3COO)_2$ in ethanol were developed in [8]. The average sizes 4 ± 0.4 nm of ZnO nanoparticles were determined being based on the absorption spectra and atomic force microscopy.

The method of synthesis in aqueous and ethanol solutions of polyethyleneimine of ultra-small cadmium sulfide nanoparticles was developed for the conditions of full saturation of the Cd cations with amino groups [9]. The average sizes of CdS nanocrystals (1.8…2 nm) were determined using atomic force microscopy. X-ray diffraction spectra of CdS nanocrystals in poly-ethyleneimine confirmed the crystalline structure of nanoparticles.

ZnO or CdS nanoparticles were deposited onto the surface of macropores from the colloidal solutions in polyethyleneimine at the following ratio: nanocrystals $10 \pm 2\%$; polyethyleneimine $18 \pm 2\%$; water – the rest.

SiO_2 nanocoatings were formed in the diffusion furnace after treatment of macroporous silicon substrates in the nitrogen atmosphere [10]. The oxide layers (thickness of 5…50 nm) were formed on macroporous silicon samples in dry oxygen for 40…60 min at the temperature close to 1050 °C. Silicon oxide layers 100- and 200-nm thick were prepared for 50 min at 1100 °C in wet oxygen atmosphere by using the steam from deionized water. The oxide thickness was measured using ellipsometry.

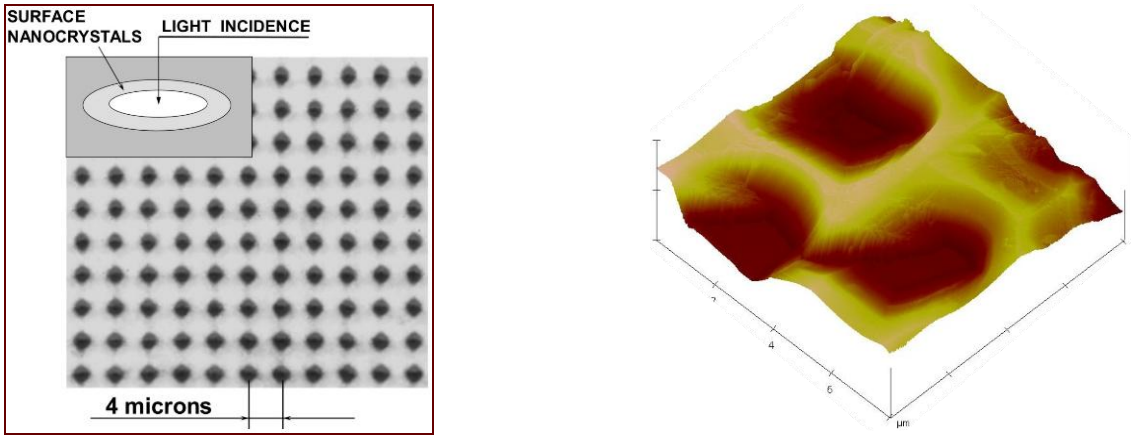
We performed optical investigations within the spectral range 300…7800 cm^{-1} by using the IR Fourier spectrometer "Perkin Elmer" (Spectrum BXII). The optical absorption spectra were measured at normal incidence of IR radiation on the sample (along the cylindrical macropores, Fig. 1a, insertion) in the air at room temperature. The error of spectral measurements was about 2 cm^{-1}.

3. Experimental

IR spectra. For macroporous silicon structures with nanocoatings and microporous silicon layers the light absorption increases, and an oscillating structure with giant amplitudes occurs (Fig. 2a).

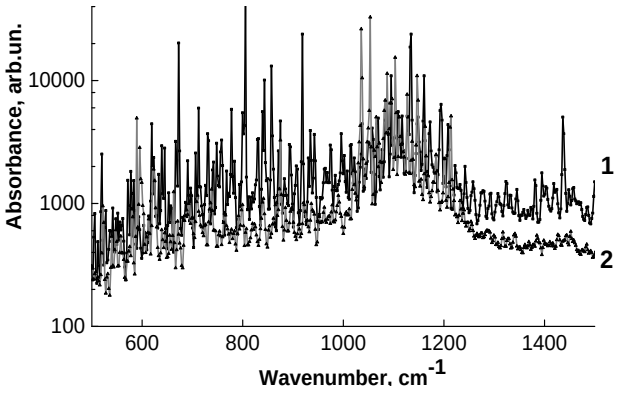
Fig. 2a shows the experimental dependences of the IR absorption in macroporous silicon structures with the SiO_2 layer 50-nm thick (*1*) and microporous layer 100-nm thick (*2*). Fig. 2b shows the experimental dependences of the IR absorption in macroporous silicon structures with surface nanocrystals CdTe (*1*), ZnO (*2*), CdS (*3*) within the spectral range 500…1500 cm^{-1}.

Fig. 2c shows the experimental dependences of the IR absorption in the macroporous silicon structures with surface ZnO nanocrystals: curve *1* – within the spectral range 500…1500 cm^{-1}; curve *2* – the same spectrum of IR absorption but moved by 2 periods.

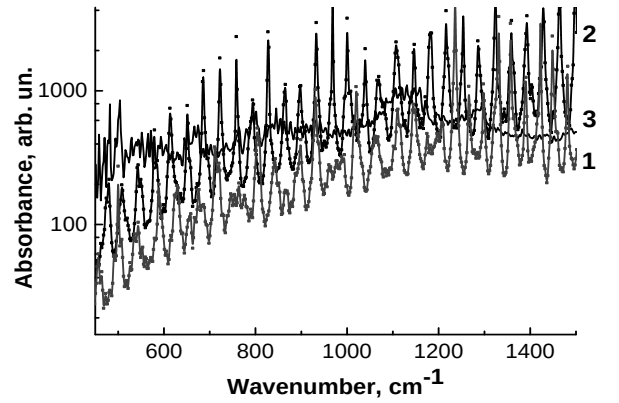


a) b)

Fig. 1. a) Macroporous silicon structures with the macropore diameter $D_p = 2$ μm and period 4 μm. Insertion: normal incidence of IR radiation on a sample (along the pores). b) Fragment of macroporous silicon structure with CdTe nanocrystals.



a)



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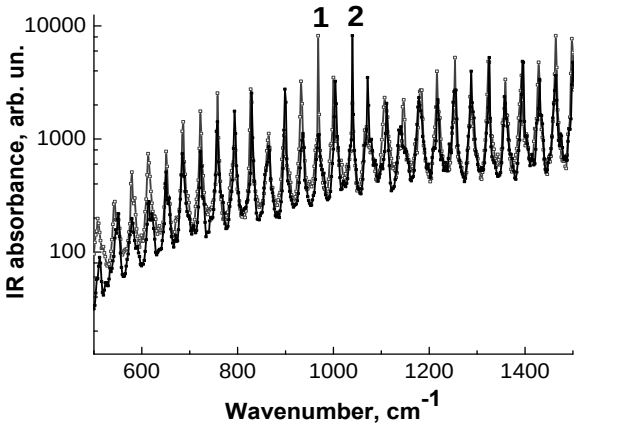

c)

Fig. 2. Experimental dependences of the IR absorption in macroporous silicon structures with (a) the SiO_2 layer 50-nm thick (*1*) and microporous layer 100-nm thick (*2*) within the spectral range 500…1500 cm^{-1}; (b) surface nanocrystals CdTe (*1*), ZnO (*2*), CdS (*3*); (c) surface ZnO nanocrystals: *1* – within the spectral range 500…1500 cm^{-1}, *2* – the same spectrum of IR absorption but moved by 2 periods.

The low coherence of oscillations is inherent to macroporous silicon with SiO_2 nanocoatings (Fig. 2a, curve *1*) and with the microporous silicon layer (Fig. 2a, curve *2*), which is caused by high concentrations of surface states [4, 10]. A better oscillation coherence was obtained for CdTe and ZnO nanocrystals (Fig. 2b, curves *1* and *2*). However, for structures of macroporous silicon with CdTe nanocrystals (Fig. 2b, curve *1*) of the size 20 nm, the coherence of oscillations is less of ZnO nanocrystals with the nanocrystal sizes of 4 ± 0.4 nm (Fig. 2b, curve *2* and Fig. 2c). The structures of macroporous silicon with CdS nanocrystals of 1.8…2 nm (Fig. 2b, curve *3*) are also significantly inferior to those of ZnO nanocrystals.

Thus, the coherence of oscillations increases with the decrease in the concentration of surface states and with the optimal contact of nanocrystals to the surface of the macropores due to optimal size of nanocrystals (4 ± 0.4 nm). For ZnO nanoparticles, the distance between resonant oscillations is 4.4 ± 0.28 meV, and the shift of oscillations by 1 and 2 periods (Fig. 2c) leads to deviation of the oscillation peaks within the range 0.26…0.42 meV, *i.e.*, the oscillation coherence reaches 0.25–0.4%.

4. Discussion

An effective mechanism for implementation of strong coherent interactions is the resonant oscillations caused by the Wannier–Stark quantum electro-optical effect in IR absorption spectra [3, 11]. The basis for developing this direction is interaction of the quantum system of oscillating electrons with the quantum system of Wannier levels. The Wannier–Stark steps can exist when the scattering time τ_s of the carriers is sufficiently large to realize at least one cycle of complete Bloch oscillation of the field-accelerated carriers (Fig. 3a). In our case, the electric field is at the boundary of the silicon matrix – nanocoating, and the vector of electric field lies in the (100) plane of silicon (Figs 3a, 3b). After illumination, electrons are accelerated in the electric field of the enriched electric potential (Fig. 3a), oscillate and are scattered by surface states in the radial direction *x* relatively to the macropore, in the *y*, *z* plane (Fig. 3b) that is the plane of resonant scattering with an infinite amplitude [11]. The resonant electron scattering results in the infinite growth of dielectric functions and corresponding changes in IR absorption at room temperature.

The Wannier–Stark steps have a certain width Γ, so this width should be less than the energy difference of the neighboring steps, $\Gamma < Fa$. The Wannier–Stark steps are not changed, if the electron scattering time by impurities τ is greater than the period of electron oscillations in the electric field T_B ($\tau/T_B > 1$, where $T_B = 2\pi h/\Delta E$). In addition, τ is proportional to $1/W$ (W– probability for electron to leave the state per time unit due to scattering by impurity atoms at the lattice nodes). The electron scattering time in macroporous silicon structures is relatively large due to the low concentration of surface impurities at the surface of macropores ($N_i \le 5 \cdot 10^{10}$ cm^{-2})

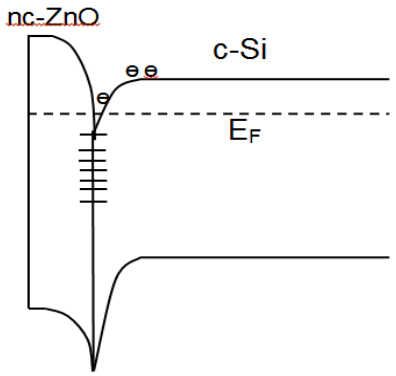

Fig. 3a. Scheme of potential well and band bending at the surface of macropores between silicon c-Si matrix with the Fermi level E_F and layer of nc-ZnO nanocrystals (circles – electrons, strokes – impurity states).

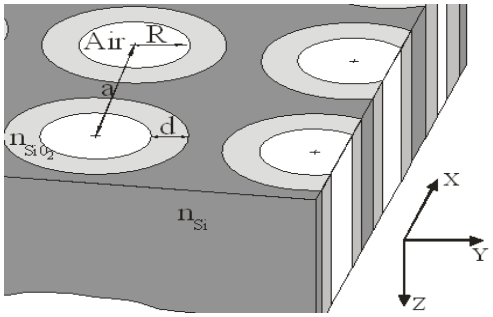

Fig. 3b. A fragment of the macroporous silicon structure with the nanocoating system considered. The potential of impurities in the electric field of a deep well (Fig. 3a) between the macropore surface and the nanocoating is sufficient.

[3, 10]. Therefore, the inequality $\tau/T_B > 1$ is satisfied within the entire region of the measured IR spectra. The structures of macroporous silicon have the density of scattering centers on the surface of macropores approximately $10^{10}…10^{11}$ cm^{-2}. The difference between these two resonant energies is 8…20 cm^{-1}, depending on the material of nanocoatings.

Controllability and coherence of the system are defined by formation of coherent Wannier levels in a narrow triangular potential well formed by an electric field at the silicon – nanocoating boundary (Fig. 3a). The presence/absence of resonance scattering of oscillating electron in the electric field with the potential of impurity surface states with quantum Wannier levels is controlled and measured at room temperature by the resonant maxima of the IR absorption. The resonant maxima are stored for 20 s after switching off the illumination. This is 200 times longer than that of macroporous silicon structures without nanocoatings [12].

Electron-phonon interaction. We investigated the contribution of the electron-phonon interaction to the broadening parameter Γ of the Wannier–Stark steps in oxidized macroporous silicon structures [13]. The effect of broadening on the amplitude of oscillations in IR absorption spectra (ΔA) is calculated in the form of convolution of the "non-broadened" oscillation amplitudes (ΔA_0) with the Lorentz distribution:

$$\Delta A/\Delta A_0 = \frac{\Gamma}{\pi}\int \frac{d\omega'}{(\omega'-\omega)^2+\Gamma^2} = \arctan(\Delta\omega/\Gamma)/\pi\,. \qquad (1)$$

In Eq. (1), we used $\Delta\omega = \omega' - \omega$ as the energy of the Wannier–Stark step Fa.

For experimental data on IR absorption by macroporous silicon structures with the oxide thickness between 10 and 200 nm in the spectral region of Si–O surface states, $\Delta A_0/\Delta A$ = 3…20 (Fig. 4), which is confirmed by low values $\Gamma/Fa << 1$, where Γ = 0.3…0.8 cm^{-11} equals to that for surface phonon-polaritons measured in thin films of II–VI semiconductors [14]. The obtained parameter Γ of the Wannier–Stark step is much less than the adjacent energy level due to the giant amplitude of oscillations after resonant electron scattering by the surface states with the infinite amplitude of scattering.

Quantum superiority. To achieve a high degree of coherence for the quantum system at room temperature, it is necessary to achieve "quantum superiority" due to the hundreds of coupled acts of the electron scattering operating steadily and with a small number of errors, high accuracy and speed of measurements.

On the other hand, the potential of impurities in the electric field of a deep well (Fig. 3a) between the macropore surface and the nanocoating is sufficient. Thus, the influence of "quantum superiority" on the coherence is significant to form the Wannier levels. Moreover, oscillating motion of electron together with its spin enhances the circulating flow of energy in the field of its wave. Resonant scattering rotates the electron flow by 90° and converts the superposition of states inherent to the microscopic system into the superposition of states of the macroscopic system. This mechanism includes also interaction of the macropore quantum system with other macropores, which causes confusing quantum correlation and forms the superposition of states in the macroscopic system. Effective formation of quantum states and the strengthening of quantum systems on the surface of periodically located macropores result in the low value of the broadening parameter Γ (10^{-3} of Wannier–Stark step width), and the oscillation coherence reaches 0.25–0.4%.

The obtained results are promising for developing the optical quantum computer [15–17]. Such a quantum computer is a computing device that uses the phenomena of quantum mechanics (quantum superposition, quantum entanglement) to transmit and process data. The quantum computer operates not with bits possessing the values of either 0 or 1, but with qubits having values of both 0 and 1. As a result, it is possible to process all the possible states simultaneously and to obtain a huge advantage over other computers in a number of algorithms [16].

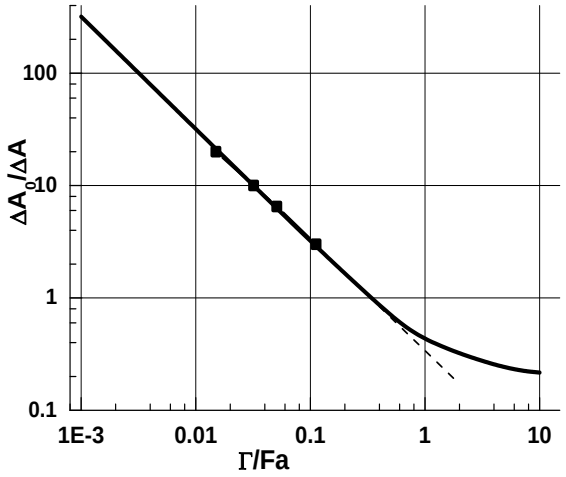

Fig. 4. Results of calculation (curve) of the $\Delta A_0/\Delta A$ dependence on Γ/Fa by using Eq. (1). The symbols are experimental data on IR absorption by macroporous silicon structures with the oxide thickness from 10 to 200 nm in the spectral region of Si–O–Si surface states.

The logical qubit in our case is the presence/absence of the resonant scattering of oscillating electrons by Wannier levels in the electric field at the boundary of the silicon matrix – nanocoating. This qubit corresponds to electrons in the quantum state of $|0\rangle$ and $|1\rangle$ in the plane of the infinite resonance scattering controlled by IR illumination and the resonant maximum of IR absorption. The number of oscillations in one IR spectrum is more than 100, thus, we have a multi-qubit system.

Interaction of qubits in arrays of individually controlled, cold neutral Ba^+ atoms [18] is propagation of internal atomic coherence to and from the direction of centers of mass motion for all ions. In this system, the total time of measurement is 35 s, which is much less than decoherence due to spontaneous emission (45 s), so the decoherence time is 6 s with an error of 5%. A two-qubit quantum computer based on a diamond crystal with impurities [19] includes two logical qubits of the electron spin and the nitrogen cores with the coherence 10–17% at room temperature. As a result, the broadening parameter 10^{-3} of the Wannier–Stark step width, the coherence of oscillations 0.25–0.4% at room temperature is much less than the coherence of cold atoms (5%) and a diamond crystal with the nitrogen impurity (10–17%) at room temperature.

The presence/absence of resonant scattering of oscillating electron in an electric field at the potential of an impurity surface state with the quantum Wannier levels is controlled at room temperature by the resonant maxima of the IR absorption, which is 200 times longer than that of macroporous silicon structures without nanocoatings [12]. Thus, we proposed the high-coherent optical quantum computer based on a macroporous silicon structure with the surface layer of nanocrystals for implementation of the Wannier–Stark quantum electro-optical effect [20].

5. Conclusions

MIR light absorption oscillations in 2D macroporous silicon with CdTe, ZnO, CdS surface nanocrystals, microporous and SiO_2 layers were compared, taking into account the electro-optical effects in strong electric fields. CdTe nanocrystals (20 nm in size) were deposited on macroporous silicon substrates using the "hot wall" molecular epitaxy. The methods of synthesis of ZnO nanoparticles (3.7…4.4 nm in size) in isopropanol and from solution of zinc compound $Zn(CH_3COO)_2$ in ethanol have been developed. Ultra-small CdS nanoparticles (1.8…2 nm in size) were prepared under conditions of full saturation of the Cd cations with amino groups in aqueous and ethanol solutions of poly-ethyleneimine. SiO_2 nanocoatings (with the thickness 5…280 nm) were obtained in the diffusion furnace after treatment of macroporous silicon substrates in the nitrogen atmosphere.

The coherence of IR spectra oscillations increases with the decrease of concentration of surface states and with the optimal contact of nanocrystals to the surface of macropores. Thus, the shift of oscillations for ZnO nanoparticles with optimal size of nanocrystals (3.7…4.4 nm) leads to deviation of the oscillation peaks within the range 0.26…0.42 meV, *i.e.*, the oscillation coherence reaches 0.25–0.4%.

The small broadening parameter of the Wannier–Stark step levels $\Gamma = 0.3…0.8\ cm^{-1}$ equals to that for surface phonon-polaritons measured in thin films of II–VI semiconductors. The potential of impurities in the electric field of a deep well between the macropore surface and the nanocoating is sufficient. Thus, the influence of "quantum superiority" on coherence is significant to form the Wannier levels. In addition, oscillating motion of electron together with its spin enhances the circulating energy flow in the field of its wave. Resonant scattering rotates electrons by 90° and converts superposition of the states inherent to the microscopic system into superposition of the states related to the macroscopic system. The gain mechanism also involves interaction of the quantum macropore system with other macropores, involving them in a mixed state. As a result, the broadening parameter 10^{-3} of the Wannier–Stark step width and the coherence of the oscillations 0.25–0.4% at room temperature are much less than the coherence of cold atoms (5%) and a diamond crystal with the nitrogen impurity (10–17%) at room temperature. Thus, we propose the high-coherent optical quantum computer based on a macroporous silicon structure with the surface layer of nanocrystals for the implementation of the Wannier–Stark quantum electro-optical effect.

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Висококогерентні осциляції в ІЧ-спектрах макропористого кремнію з нанопокриттями

Л.А. Карачевцева, О.О. Литвиненко

Анотація. Одним з перспективних матеріалів для розробки двовимірних фотонних структур є макропористий кремній, отриманий за допомогою фотоанодного травлення. Наявність періодично розташованих циліндричних пор, розділених кремнієвими колонами, забезпечує велику ефективну поверхню структур, покращує оптичні та фотофізичні характеристики макропористого кремнію. У даній роботі досліджено осциляції ІЧ-поглинання двовимірними структурами макропористого кремнію з мікропористими шарами кремнію, SiO_2 нанопокриттями та CdTe, ZnO поверхневими нанокристалами з урахуванням електрооптичного ефекту Ваньє–Штарка. Аналіз експериментальних спектрів поглинання здійснено в рамках моделі резонансного розсіювання електронів з нескінченною амплітудою на поверхневих станах у сильному електричному полі, з різницею між двома резонансними енергіями, що дорівнює сходинці Ваньє–Штарка. Постійність періоду осциляцій вказує на реалізацію ефекту Ваньє–Штарка на довільно розподілених поверхневих станах на межі «кремній–нанопокриття». Проведено порівняння ІЧ поглинання світла в 2D структурах макропористого кремнію з поверхневими нанопокриттями, проаналізовано зрушення і відхилення вершин коливань. Когерентність осциляцій ІЧ-спектрів підвищується в результаті зменшення концентрації поверхневих станів та оптимальної площі контакту нанокристалів до поверхні макропор. Таким чином, зсув осциляцій для наночастинок ZnO з оптимальним розміром нанокристалів (3,7…4.4 нм) призводить до відхилень коливань у межах 0,26…0,42 меВ, тобто узгодженість коливань досягає 0,25…0,4%. Малий параметр уширення сходинок Ваньє–Штарка $\Gamma = 0{,}3…0{,}8$ см$^{-1}$ дорівнює цьому параметру для поверхневих тонких плівок напівпровідників II-VI. Контрольованість і когерентність дослідженої системи визначається формуванням когерентних рівнів Ваньє у вузькій трикутній потенціальній ямі, сформованій в електричному полі на границі «кремній–нанопокритя». У результаті було запропоновано висококогерентний оптичний квантовий комп'ютер на основі реалізації квантового електрооптичного ефекту Ваньє–Штарка на кремнієвій матриці з макропорами та шаром нанокристалів на поверхні макропор.

Ключові слова: макропористий кремній з нанопокриттями, нескінченне резонансне розсіювання, оптичний квантовий комп'ютер.